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On the role of initial conditions in the selection of eutectic onset mechanisms in directional growth

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Abstract

The early-stage dynamics and onset mechanisms for eutectic solidification are investigated experimentally using slab-geometry slides of succinonitrile–(D)camphor (SCN–DC) transparent organic eutectic material. By specifically focusing separately on the pre-growth or holding period and the growth or pulling period, the critical roles of each in the establishment of initial conditions and the competition between eutectic initiation mechanisms, leading to the development of a steady-state eutectic front, are examined. It is found that a single-phase layer forms and increases in thickness monotonically with time during the holding period with a corresponding increase in the interface temperature. Because the thickness of this layer is observed to influence subsequent eutectic initiation mechanisms, it is concluded that the pre-existing structure, holding period duration, single-phase identity and thickness, and specimen slide geometry should all be reported as standard practice, along with the pulling velocity and thermal gradient, for a complete description of a gradient-zone directional solidification experiment.
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1. Introduction

Multiphase solidification morphologies are very common in nature and are frequently observed to be major constituents in the microstructures of cast components. Eutectic structures in particular, which themselves may vary considerably in appearance [1–5], are commonly found in Fe-, Al-, Cu- and Mg-based alloy castings [6]. Beyond these conventional examples, eutectic solidification is very important in the development of new materials for which periodic multiphase structures may give rise to remarkable or enhanced functionality, such as nanoscale metallic glass/crystalline composites [7–10] and organic photovoltaics/LEDs [11,12]. The growth of coupled two-phase structures has been a principal topic of investigation for several decades [13], and while steady-state eutectic growth has been studied extensively [14] and is reasonably well understood, there has been far less attention given to the early-stage dynamics associated with the initial formation of the eutectic structure. Recently reported investigations [15–18] have shown, however, that the understanding of these onset dynamics may be critical to the effective control of array order in eutectic and other multiphase solidification microstructures, having direct implications regarding best practices for experimental methodologies and new solidification processes.

Much of our current fundamental understanding of eutectic solidification arises from the analytical treatment of Jackson and Hunt (JH) [19] and numerous experimental investigations which have generally verified the JH treatment. Beyond these simple descriptions, several other more complex steady-state or oscillatory growth morphologies have been observed experimentally [1,15,20–23] and described theoretically [24–28]. Most recently, the investigation of eutectic growth in ternary and quaternary systems has revealed a multitude of complex morphologies

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which remain to be understood [29–39]. One prevailing question that surrounds all of the experimental results, however, is related to the effects of the initial conditions that may exist in a Bridgman-type gradient-zone directional solidification (DS) experiment, which is the method through which most of the systematic experimental data have been obtained. Specifically, it has not been standard practice to carefully characterize the initial conditions in such an experiment. Rather, it has generally been assumed, perhaps without due cause, that these effects are of little importance with respect to the ultimate steady-state growth mode. Here, we examine this issue more carefully.

As we have reviewed previously [17,18], transparent metallic analog organic systems have been instrumental in the investigation of the dynamics of eutectic solidification, since they allow in situ observation of morphological evolution. These materials are generally studied using thin slide or tube geometries, where a typical simple DS experiment involves the establishment of an eutectic growth front by translating the specimen with respect to an imposed thermal gradient frame. While specimen preparation methods and ampoule geometry are routinely reported, little attention is typically given to the detailed method by which the steady-state condition is established. In particular, the initial conditions that arise from the prior microstructure and the stationary holding period have not generally been controlled or even reported. However, our previous work [18] has clearly shown that the early-stage dynamics that lead to the onset of eutectic growth under DS conditions may depend critically on these experimental details and on the specimen geometry.

In the simplest case, for a constant thermal gradient and fixed specimen geometry, the initial conditions and early-stage dynamics are dictated by three experimental factors. These are (i) the prior directional growth velocity and duration, (ii) the duration of the stationary holding after the prior growth but before the experimental growth period (iii) the pulling velocity and duration used for the experimental growth period. In the work presented here, we are interested in the evolution of structure during these latter two stages, which we define as the holding period and the pulling period, respectively.

During the holding period, the prior eutectic structure evolves considerably, and the coarsening and dissolution processes that occur here have been observed to lead to the formation of a single-phase (SP) layer between the prior eutectic (PE) structure and the liquid (L) [15,16,18,40,41]. The formation of such a layer has typically been attributed to deviation from the eutectic composition. Indeed, the invariant nature of the three-phase equilibrium at fixed pressure does not allow for self-regulation and establishment of a stable stationary boundary as would be expected along the liquidus of one phase, where the position of the interface can adjust itself within the experimental gradient frame to accommodate slight deviations in composition. In the case of the eutectic front at fixed pressure, both the temperature and composition required for equilibrium are fixed. While the interface can move within the temperature field to locate the invariant temperature, the composition in the sample is not free to adjust. Moreover, the specific eutectic composition is infinitely narrow such that it can never be attained exactly in practice. Accordingly, it is not surprising that the observation of SP layer formation is rather general. In addition, it is worth noting that stabilization of the three-phase (eutectic–liquid) interface in a thermal gradient would require the Gibbs–Thomson condition to be satisfied simultaneously at every location along the solid–liquid interface for each phase and everywhere along the included phase boundary groove, in a manner similar to a grain boundary groove [42]. Moreover, a stable condition would require equilibrium at the triple junction. Clearly, the issue of stability of the eutectic–liquid front in a thermal gradient deserves specific attention. However, we leave this subject for later analysis and simply state here that there is no justification for assuming a priori that a static solution exists in general. Instead, we simply recognize that the structure in this vicinity is complex and that it evolves through active coarsening processes. It should also be mentioned here that there are two reported studies [15,43] in which only a discontinuous SP layer was observed. We point out, however, that these experiments utilized holding periods of only 2 h, which is considerably shorter than the holding periods we employ in the present study.

During the pulling period, the eutectic onset may occur through a number of mechanisms involving the PE structure, the single-phase layer and the SP/L interface. Most simply, the early-stage dynamics involve a competition between (i) a seeding mechanism, where single-phase or eutectic growth may originate within the SP layer or at the SP/PE interface and rapidly advance toward the SP/L interface, and (ii) an interfacial mechanism involving nucleation at the SP/L interface, where grain boundary structures and specimen geometry may be important. Fig. 1 illustrates this competition. Upon pulling, the SP/L interface advances in the material frame but retreats in the thermal gradient frame, moving toward lower temperatures. While the SP front is moving, a single-phase or eutectic "seed" that formed in a grain boundary channel some time after initiation of the pulling period advances in the gradient frame. When the advancing seed reaches the SP/L interface, several mechanisms for the onset of eutectic growth may rapidly ensue.

We have shown [18] that the competition between onset mechanisms can be strongly influenced by specimen thickness (δ), where the through-thickness curvature of the SP/L interface may reduce the required travel distance for the advancing seed to reach the growth front. Indeed, while multiple mechanisms were observed in thin ($\delta = 20\ \mu m$) slides, eutectic onset was observed only through the seeding mechanism in thick ($\delta = 200\ \mu m$) slides. In addition, the seed itself was observed to be single-phase (perhaps divorced eutectic) in the thin specimens but of eutectic character in the thick specimens, as shown in Fig. 2. This

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